

Effect of disorder on the Kondo behavior of thin Cu(Mn) films

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Abstract

We have studied the influence of disorder on the Kondo effect in thin films of Cu(Mn), i.e., Cu doped with a small amount of Mn. We find that the Kondo contribution to the resistivity is suppressed when the elastic mean-free-path, λ , is reduced. While this is qualitatively similar to results found previously by our group in a different material, our new experiments reveal in detail how this suppression depends on both film thickness and λ . These results are compared with the theory of Martin, Wan, and Phillips. While there is general qualitative agreement with this theory, there appear to be some quantitative discrepancies.

I. INTRODUCTION AND BACKGROUND

The behavior of magnetic impurities in metals, and in particular the Kondo effect [1], has been of interest for several decades [2]. Even so, it is the object of much current work, as a number of important issues in this area remain unresolved. One of these issues is the Kondo behavior in small structures; that is, in thin films and narrow wires. Several years ago, experiments [3–10] revealed that the Kondo effect can depend on system size. The Kondo effect makes a contribution to the resistivity which, at high temperatures, has the form

$$\Delta\rho_K = -B_K \ln(T) . \quad (1)$$

The conventional Kondo effect leads to an increase in the resistivity at low temperatures ($\Delta\rho_K > 0$), so the coefficient B_K is positive. The experimental studies noted above concerned $\Delta\rho_K$ in thin films and narrow wires, and found that B_K becomes smaller when the system size is reduced; that is, the Kondo effect is suppressed in small systems [11]. These experiments also revealed that B_K depends on the level of disorder, with its value decreasing as the elastic mean-free-path, λ , is made smaller. Quite recently, a similar size dependent suppression of the Kondo effect was observed in the thermopower [15].

Qualitatively, the Kondo effect is due to the screening of a magnetic impurity by the conduction electrons [1,2], and it is the interaction responsible for this screening which leads to (1). It was initially suggested [4] that the observed size dependence of B_K might be related to the spatial extent of the conduction electron screening cloud. However, subsequent experiments [8,10] showed that this explanation cannot be correct, as the length scale associated with the suppression does not vary with T_K in the manner expected from this general picture. In addition, it has been argued on theoretical grounds that screening cloud physics cannot explain the observed size dependence of $\Delta\rho_K$ [16,17]. While the experiments did not suggest the mechanism responsible for the size dependence, they did reveal that $\Delta\rho_K$ can also be suppressed by an increase in the level of disorder, i.e., a reduction of the elastic mean-free-path, λ .

The observations that $\Delta\rho_K$ depends on the system size and on λ have recently been addressed by two theoretical studies. An explanation for the size dependence in the clean (large λ) limit has been developed by Zawadowski and co-workers [14]. According to their picture, a combination of spin-orbit scattering and scattering from a surface (both involving the conduction electrons) gives rise to a uniaxial anisotropy at a magnetic impurity. This anisotropy has the form DS_z^2 , where D is a function of material parameters (such as the density of states) and distance from the surface. The precise value D is very difficult to estimate, so the theory cannot be judged or tested based on its prediction of the precise value of D . However, we have recently tested this theory in another way. In the experiments mentioned above, the magnetic species had integer spin; in most cases it was Fe, which is believed to be described by an effective spin $S = 2$ [18]. In this case the anisotropy energy splits the Fe sublevels, leaving a singlet $S_z = 0$ at an energy D below the $S_z = \pm 1$ doublet. When D is sufficiently large compared to $k_B T$, only this singlet ground state will be occupied, and the Fe will be nonmagnetic. This is the origin of the suppression of B_K ; as a system is made smaller, an increasing fraction of the local “moments” are near the surface, and are thus rendered nonmagnetic by the splitting arising from this anisotropy. In recent work, we investigated the behavior of a magnetic impurity with half-integer spin, Mn which has $S = 5/2$. In this case the ground state will always be a doublet, and hence magnetic. Hence, the theory predicts that in this case B_K will not vanish as the system size $\rightarrow 0$, and this was indeed observed in our experiments with Cu(Mn) [19].

In this paper, we again consider the behavior of Cu(Mn), but now we focus on the behavior of B_K as a function of disorder. In the case of strong disorder, Martin, Wan, and Phillips [20] have shown theoretically that there is an interplay between Kondo physics and the quantum interference effects which are responsible for weak localization [21]. This interplay gives B_K the form

$$B_K = B_K^0 \left(1 - \frac{\alpha}{t\lambda^2\tau_s} \right) , \quad (2)$$

where B_K^0 is the value found in bulk systems, t is the film thickness, λ is the elastic mean

free path, τ_s is the spin scattering time, and $\alpha = 1.2\hbar/\pi m k_F$ is a parameter which depends only on Fermi surface properties and is not expected vary with disorder. This result was obtained with the assumption that $\lambda < t < L_\phi$, where L_ϕ is the electron phase coherence length (which will be discussed further below) [22]. Martin, Wan, and Phillips have shown that the prediction (2) gives a good account of previous experimental results from our group for Cu(Fe) films [20].

In the present paper we present results for the behavior of B_K for Cu(Mn) as a function of λ and t , along with independent measurements of τ_s via weak localization experiments. This has allowed us to test the prediction (2) in detail; we will see that it does not seem to provide a very good quantitative account of our new results.

II. EXPERIMENTAL METHOD

Cu(Mn) has been studied a great deal in connection with the Kondo effect in bulk alloys [2], making it an ideal choice for the present experiments. From previous work, we know that a Mn concentration, c , in the neighborhood of a few hundred parts per million (ppm) should be low enough to observe behavior in the dilute limit; i.e., $\Delta\rho_K/c$ should be independent of c [23]. In order to produce films with high disorder we employed DC sputtering. To obtain Cu films with a small concentration of Mn impurities, several small pieces of manganin wire (approximately 2 mm in length and 0.5 mm diameter) were placed uniformly on the surface of a pure (99.999%) Cu sputtering target of diameter 5 cm. Manganin wire has the composition 86% Cu, 12% Mn, and 2% Ni and thus allows us to sputter a small amount of Mn along with the Cu. Using this method has two slight drawbacks. (1) the films will contain a small amount of Ni. However, since Ni does not have a local moment when placed in Cu, we do not expect its presence to affect our results [24]. (2) It is difficult to know, with great precision, the Mn concentration. This uncertainty is due to the fact that the target does not sputter uniformly over long periods of time, and the different materials have different sputtering rates, etc. We have dealt with this problem in two ways. First, we will

make direct comparisons only between samples prepared in the same sputtering session. In a single such session we deposited a series of films, with different thicknesses. This should produce films with the same Mn concentration, but with different thicknesses as determined by the time they are exposed to the sputtering beam, and different disorder as determined by the Ar pressure. Second, during each sputtering session the first and last films that were prepared were designed to have the same mean-free-path and thickness. We found that they always exhibited the same behavior (to within the uncertainties), which demonstrates that the Mn concentration was indeed constant throughout the session.

Based on the amount of manganin wire placed on the target and the relative sputtering rates of Cu and Mn, we estimate the concentration to be in the neighborhood of, or somewhat less than, 300 ppm. With the added knowledge that the data show no sign of magnetic ordering or coupling effects [25], we are confident of being in the dilute impurity regime.

In a sputtering session, a collection of substrates (glass) was mounted on a rotating holder system in such a way that only one substrate at a time was exposed to the sputtering beam. The sputtering took place with the substrates nominally at room temperature, with Ar pressures in the range 1.5 to 10 mTorr. After depositing a film, the sample holder was then rotated without breaking vacuum to expose another substrate, and the Ar gas pressure was adjusted to change the level of disorder in the next film, with higher pressures yielding films with greater disorder (i.e., shorter λ). This process was performed to make typically six films using this target setup. As noted above, the first and last films were made at the same Ar pressure as a check on our procedure.

Immediately after removing a given batch of films from the vacuum chamber they were coated with photoresist, and patterned with optical lithography and etching in dilute nitric acid, to produce strips of width $\sim 150 \mu\text{m}$ and length 60 cm. Note that the photoresist was then allowed to remain on the Cu(Mn) films, thus protecting them from oxidation. Resistance was measured as a function of temperature using a standard 4-wire DC method in a ^4He cryostat. Magnetoresistance measurements were made using an AC bridge technique with the reference resistor either at room temperature or at the same temperature as the

sample.

The Kondo temperature of Cu(Mn) is not known precisely. Previous work has established only that it is below $\sim 10^{-2}$ K. This well below the range studied here, so we will always be in the regime where the high temperature limit (1) is applicable.

III. RESULTS AND DISCUSSION

Some typical results for the resistivity as a function of temperature are shown in Fig. 1. Here we have plotted just the *change* of the resistivity with temperature, with the zero of $\Delta\rho$ chosen at a convenient temperature (here near 6 K). Below about 4 K it is seen that $\Delta\rho$ varies approximately logarithmically with T , as expected from (1). At higher temperatures (not shown here) the resistivity increases with increasing T , due to the usual effect of electron-phonon scattering. To avoid having to deal with this effect, we will restrict our attention to the behavior below about 4 K, where electron-phonon scattering is negligible compared to the Kondo contribution to $\Delta\rho$.

While the logarithmic variation seen in Fig. 1 is quite consistent with (1), there are two other effects which can give rise to a similar temperature dependence. In two dimensions, electron-electron interaction effects (EEI) [26] give rise to a logarithmic variation of the sheet resistance, R_{\square} , which has the form

$$\frac{\Delta R_{\square}}{R_{\square}} = -\frac{e^2}{2\pi^2\hbar} A_{ee} R_{\square} \ln T , \quad (3)$$

where A_{ee} is a screening factor which is typically near unity in metal films [26,27]. Because of the dependence on R_{\square} , this contribution is much smaller than the Kondo effect for large thicknesses. So far as we know, it is believed that the Kondo and EEI contributions are additive. With this assumption, we have subtracted the contribution calculated from (3) from our measurements (this subtraction was performed also for the data in Fig. 1). The screening factor A_{ee} was obtained from careful measurements with pure Cu films (prepared under the sputtering conditions given above) with resistivities and thicknesses similar to

the Kondo samples of interest. This was accomplished by measuring the variation of the resistivity (or equivalently, the sheet resistance, since the thickness was known) with temperature. By using pure Cu films, there was no Kondo contribution, and by restricting the measurement to low temperatures (below about 4 K) the electron-phonon contribution discussed above was negligible. This left only the EEI effect and weak localization (WL), which we will discuss in detail in a moment. The WL effect was quenched (without affecting the EEI contribution) by the application of a magnetic field of 15 kOe perpendicular to the film plane. From the results for several samples we found $A_{ee} = 1.0 \pm 0.1$, a value which is quite in line with that reported previously for similar films [27]. The uncertainty in A_{ee} is a conservative estimate which encompasses all of our results for Cu(Mn) films over a wide range of thickness. This value for A_{ee} will be used below to subtract the EEI contribution from the total temperature variation found for our Cu(Mn) films.

Another contributor to our measured temperature dependence is weak localization (WL). This is a quantum interference effect [21] which makes a contribution of the form

$$\frac{\Delta R_{\square}}{R_{\square}} = \frac{e^2}{4\pi^2\hbar} R_{\square} \ln L_{\phi}(T) , \quad (4)$$

for our Cu and Cu(Mn) samples (which exhibit antilocalization in small fields). Here L_{ϕ} is the electron phase coherence length. If, as happens to occur in many cases, L_{ϕ} varies as a power of T , the WL contribution varies logarithmically with temperature, with a magnitude similar to that of the EEI effect. However, in Cu(Mn) the phase coherence length is limited by the effect of the process of spin scattering [21], in which the spin of a conduction electron is flipped through an interaction with the local moment (Mn). This typically gives rise to phase coherence length which varies only weakly, if at all, with temperature [28].

In most cases below we are not interested in the WL contribution. For our thickest samples, which have the smallest sheet resistances, the WL effect is generally negligible (since the WL effect (4) is proportional to R_{\square}). It becomes more important for the thinnest films, but even in such cases it can be avoided by performing the measurement in a magnetic field. As noted above, a large magnetic field applied perpendicular to the film quenches WL. While

WL can thus be easily avoided, it can also provide some important information. The theoretical prediction (2) involves the spin scattering time, τ_s . When the phase coherence length is dominated by spin scattering, it is given by $L_\phi = \sqrt{D\tau_s}$, where $D = v_F\lambda/3$ is the electron diffusion constant (here v_F is the Fermi velocity). Measurement of the magnetoresistance in low fields, which is due solely to WL, can be used to extract L_ϕ and hence also τ_s . We will make use of this fact below.

Returning to Fig. 1, we see that varying the level of disorder, i.e., λ , has a large effect on the behavior. This can also be seen from Fig. 2, which shows results for B_K as a function of λ , for different values of film thickness, t . Each data point was obtained from a measurement of the resistivity, ρ , as a function of T , like the ones shown in Fig. 1. The results for $\rho(T)$ were fit to a logarithmic form, and the EEI effect was subtracted using the (previously) measured value of A_{ee} discussed above. Note also that we give results for magnetic field $H = 0$, and for $H = 5$ kOe applied perpendicular to the plane of the sample; the latter should be sufficient to quench WL. Typical uncertainties are shown; they are seen to become larger for the thinner films, for the following reason. As the film thickness decreases, the sheet resistance increases, making EEI larger relative to the Kondo effect. Our uncertainty in A_{ee} then leads to a larger uncertainty in B_K . The solid curves in Fig. 2 are simply guides to the eye drawn through the data for $H = 5$ kOe (the filled symbols). The dashed curves have the general function form predicted by the theory (2).

Before discussing the results in Fig. 2 we should note again that all of the data in a given plot were obtained from samples prepared in a single sputtering session, and hence had the same Mn concentration. However, this concentration varied somewhat from one session to the next, so differences in the absolute scale of B_K in the different cases are likely due, completely or in large part, to variations in the concentration. This should not affect either the general shapes of these curves as a function of λ , or the variation with field, and these are what we will rely on in our analysis below.

The thickest samples, with $t = 700$ and 400 Å, exhibit similar behavior. For large disorder, i.e., small λ , B_K appears to approach zero to within the uncertainties, except

perhaps for the smallest values of λ where the uncertainties (due to uncertainties in the EEI subtraction) become very large. As λ is increased, B_K also increases, with this increase becoming more rapid when λ exceeds a “threshold” value. This threshold is $\lambda \sim 500$ Å for the samples with $t = 700$ Å, and decreases to $\lambda \sim 150$ Å when the thickness is reduced to 400 Å. It is especially noteworthy that the behavior for these two values of the film thickness is affected very little by the application of a magnetic field. The only change is a decrease in B_K in the presence of a field; the magnitude of this decrease is very small for the case of weak disorder (large λ) and becomes larger as λ is reduced. Qualitatively and quantitatively, it would appear that the only effect of a field is to quench weak localization.

The behavior for the thinner samples is more difficult to determine, as the uncertainties are larger (for the reasons discussed above). The results for the 275 Å thick samples are consistent with $B_K \rightarrow 0$ as λ becomes small, or with a B_K which is independent of λ . The uncertainties for the $t = 150$ Å samples are also substantial. Here again the data are consistent (barely) with B_K being independent of disorder, although they seem to prefer a value of B_K which grows substantially as $\lambda \rightarrow 0$.

Let us now compare these results to the theory (2). Taken at face value, (2) would seem to predict that B_K can become negative, i.e., a “negative” Kondo effect, when either λ is made sufficiently short, or the thickness t is made very small. To within our uncertainties, we have no evidence for a negative B_K in these limits. However, it seems quite plausible that the expression (2) cannot be extrapolated to the regime where $\alpha/(t\lambda^2\tau_2) \sim 1$; i.e., other higher order terms or contributions may then be important, etc. If this is the case, then (2) cannot be meaningfully extrapolated to the parameter regime where it yields a negative B_K . However, it may still make sense to use it to estimate the “threshold” values of λ noted above. That is, the values of λ at which B_K is observed in Fig. 2 to increase substantially may be estimated from the condition

$$\frac{1}{t\lambda^2} \approx constant , \quad (5)$$

which is obtained by simply setting (2) to zero. The condition (5) suggests that the threshold

value of λ should increase as the thickness is reduced. However, this is opposite the trend observed in Fig. 2, as the thickness is reduced from 700 to 400 Å. It does not appear that the trend found in our experiment can be accounted for by (2).

The effect of a field on B_K is also noteworthy. The disorder correction to the bulk Kondo effect in (2) arises from a Kondo contribution to the spin scattering time which then affects WL. According to this theoretical picture, application of a magnetic field should not only quench the “ordinary” WL effect, but also destroy the suppression of B_K . This means that a field should cause the Kondo contribution to *increase* with respect to its value in zero field. Such behavior is contrary to what is observed in our experiments, Fig. 2, where B_K is seen to be either essentially constant, or decrease, in the presence of a field.

A. Spin scattering rate

The spin scattering time, τ_s , plays a key role in the prediction (2). We have therefore used measurements of the WL magnetoresistance to obtain an independent measure of τ_s . Typical results for the variation of the resistivity as a function of magnetic field applied perpendicular to the film plane are shown in Fig. 3. The analysis of such measurements are now standard, and are described in detail elsewhere [10,29]. A fit to the theoretical form for the WL magnetoresistance yields the phase coherence length, L_ϕ . The results from the data in Fig. 3 are 1500 Å for the 150 Å thick sample, and 1300 Å for the thicker sample, with uncertainties of approximately 100 Å in both cases. These values are in good accord with results found previously for other samples in which spin scattering was dominant [10,30]. In addition, to within the uncertainties just noted, the value of L_ϕ changed very little in going to 4.2 K, which is also expected when spin scattering is dominant.

To obtain τ_s from the phase coherence length, we must estimate the value of the diffusion constant. *If* we use nearly-free-electron theory to obtain the Fermi velocity [31], we find $v_F = 1.6 \times 10^8$ cm/s, which for the samples in Fig. 3 leads to $D \sim 15$ cm²/s. Using this value together with the phase coherence lengths found from the magnetoresistance gives

$$\tau_s \sim 1.5 \times 10^{-11} \text{ s.}$$

Let us now compare this with the theory by using (2) to calculate what value of τ_s would be needed to suppress B_K to zero at the values of t and λ observed in the experiment, Fig. 2. From the results for $t = 400 \text{ \AA}$, we estimate that $B_K \rightarrow 0$ at $\lambda \approx 150 \text{ \AA}$. Inserting these values into (2) we find $\tau_s \sim 3 \times 10^{-9} \text{ s}$ [32]. This is approximately two orders of magnitude larger than the measured τ_s . The measured value of τ_s is derived from the phase coherence length, and therefore depends on the value employed for the diffusion constant, which contributes some uncertainty. However, we do not believe that this uncertainty amounts to two orders of magnitude. The value of τ_s required to make the theory (2) compatible with our results is difficult to reconcile with our direct measurement of the spin scattering time.

IV. CONCLUSIONS

We have found that the Kondo effect in thin Cu(Mn) films is suppressed as the level of disorder is increased. This dependence on disorder is similar to that found previously for Cu(Fe). However, the present results are much more detailed, and are the first to reveal the detailed dependence of B_K on the film thickness, elastic mean-free-path, and magnetic field. The theory of Martin, Wan, and Phillips provides only a qualitative account of our data. There are several, potentially serious, quantitative discrepancies which remain unresolved, and which suggest that the crucial physics for this problem is not yet accounted for.

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FIGURES

FIG. 1. Temperature dependence of the resistivity of several 400 Å thick films. The mean-free-paths, λ , are indicated. Note that here we have plotted the *change* of the resistivity, as discussed in the text, and the magnetic field here was zero.

FIG. 2. Variation of the Kondo slope, as defined in (1), as a function of λ for different film thicknesses, t , as indicated. The open circles were obtained in zero magnetic field, while the filled circles are data obtained with $H = 5$ KOe. The difference between the two cases becomes smaller as λ is increased, and for the thickest films, $t = 400$ and 700 Å, the two results are in many cases indistinguishable for large λ . The solid and dashed lines are guides to the eye, and are discussed in the text.

FIG. 3. Variation of the resistivity as a function of magnetic field of two Cu(Mn) samples. The temperature was 1.4 K, and the sample thicknesses are indicated in the figure. The solid line for the 150 Å sample shows a fit to the theoretically expected WL magnetoresistance.

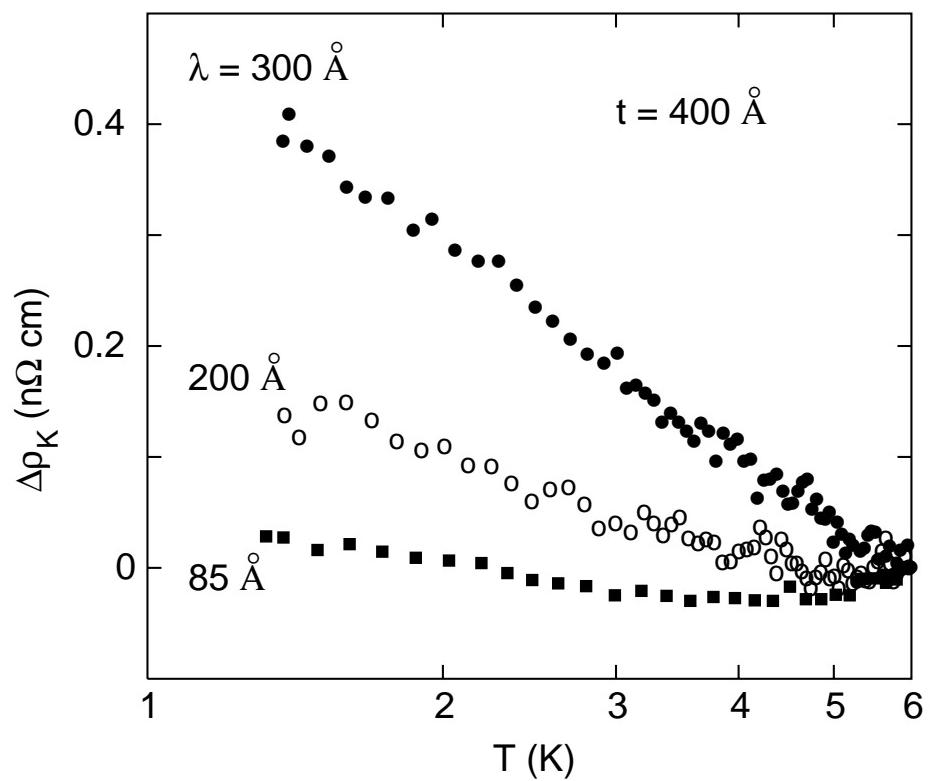


Figure 1

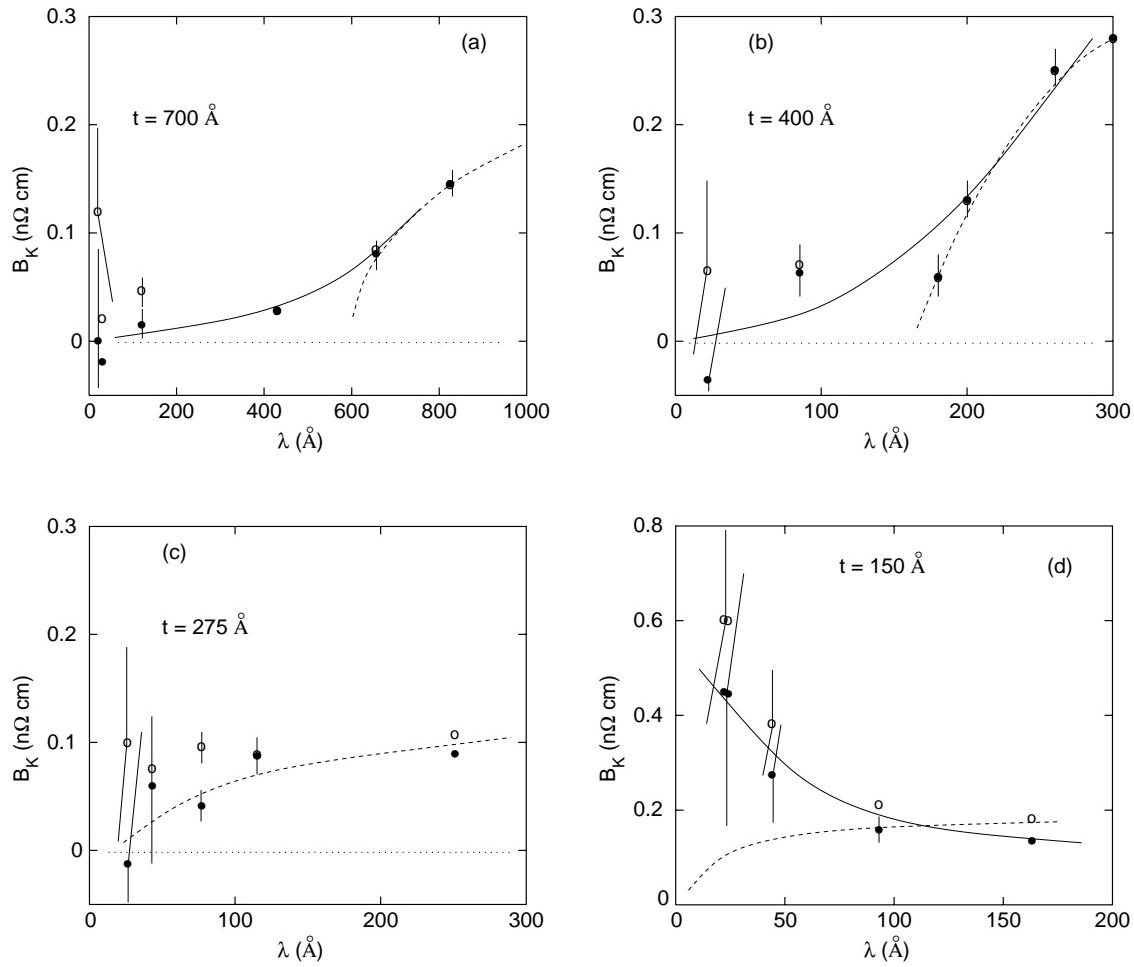


Figure 2

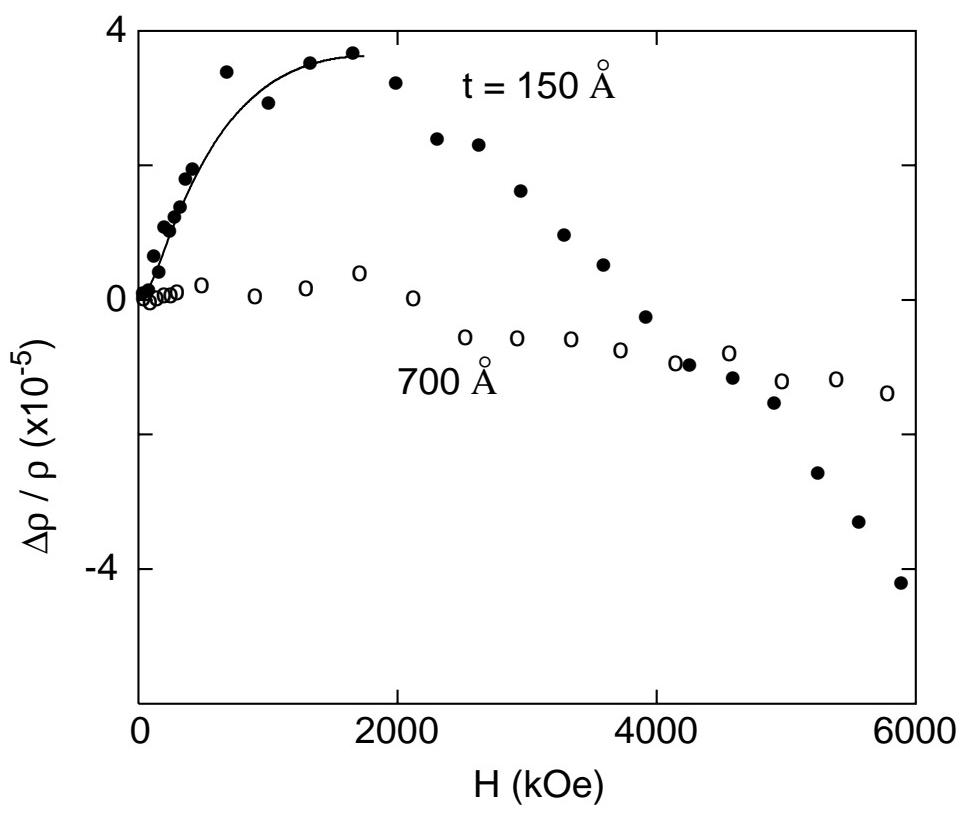


Figure 3